

Magnetism and unconventional superconductivity in $\text{Ce}_n\text{M}_m\text{In}_{3n+2m}$ heavy-fermion crystals

J. D. Thompson ^{a,1}, M. Nicklas ^a, A. Bianchi ^a, R. Movshovich ^a, A. Llobet ^a, W. Bao ^a,
A. Malinowski ^a, M. F. Hundley ^a, N. O. Moreno ^a, P. G. Pagliuso ^a, J. L. Sarrao ^a,
S. Nakatsuji ^b, Z. Fisk ^b, R. Borth ^c, E. Lengyel ^c, N. Oeschler ^c, G. Sparn ^c, F. Steglich ^c

^aLos Alamos National Laboratory, MS K764, Los Alamos, NM 87545 USA

^bNHMF, Florida State University, Tallahassee, FL 32310 USA

^cMax-Planck-Institute for Chemical Physics of Solids, Dresden, D-01187 Germany

Abstract

We review magnetic, superconducting and non-Fermi-liquid properties of the structurally layered heavy-fermion compounds $\text{Ce}_n\text{M}_m\text{In}_{3n+2m}$ (M=Co, Rh, Ir). These properties suggest d-wave superconductivity and proximity to an antiferromagnetic quantum-critical point.

Key words: $\text{Ce}_n\text{M}_m\text{In}_{3n+2m}$; d-wave superconductivity; quantum criticality

The report [1] of pressure-induced superconductivity in the heavy-fermion compound CeRhIn_5 , with a transition temperature exceeding 2 K, has motivated further exploration [2,3,4,5] of this compound and the broader family of materials $\text{Ce}_n\text{M}_m\text{In}_{3n+2m}$, where M is a transition metal Co, Rh or Ir. Diffraction studies [6,7] show that the family can be considered a structural hybrid of CeIn_3 and ' MIn_2 '; for $n = 1$, single layers of CeIn_3 and ' MIn_2 ' are stacked sequentially along the tetragonal c-axis, and for $n = 2$ there are two adjacent layers of CeIn_3 separated by a single layer of ' MIn_2 '. Crystallographic layering in the $n = 1$ members produces electronic anisotropy, reflected particularly in a Fermi surface dominated by a slightly warped cylindrical sheet. [8,9,10,11] Though these materials should not be considered strictly 2-dimensional, their electronic and structural anisotropies do influence magnetic, superconducting and quantum-critical properties. In the following, we briefly review what has been learned about some of these properties.

The infinite-layer (cubic), parent of this family, CeIn_3 , orders antiferromagnetically near 10 K at atmospheric pressure. Applying pressure suppresses its Néel temperature toward $T = 0$ at a critical pressure $P_c \approx 2.6$ GPa, where a 'dome' of superconductivity appears in a narrow pressure window centered around P_c . [12] The single and bilayer members with M=Rh also order antiferromagnetically and become pressure-induced superconductors, but both with nearly an order of magnitude higher T_c [1,5] than the maximum of ~ 0.25 K found in CeIn_3 . Some magnetic prop-

	T_N (K)	Q (h,k,l)	μ_o (μ_B)	P_c (GPa)
CeIn_3	10.2	$(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ [13,14]	0.65 [13] 0.48 [14]	2.6 ± 0.1 [12]
CeRhIn_5	3.8 [1]	$(\frac{1}{2}, \frac{1}{2}, 0.297)$ [15]	0.37 [15]	1.6 ± 0.1 [1]
Ce_2RhIn_8	2.8 [2] 1.65 [17]	$(\frac{1}{2}, \frac{1}{2}, 0)$ [16]	0.55 [16]	3 ± 0.5 [5] 0.04 ± 0.01 [5]

Table 1

Magnetic properties of $\text{Ce}_m\text{Rh}_n\text{In}_{3m+2n}$ and CeIn_3 . T_N : Néel temperature; Q: antiferromagnetic propagation vector; μ_o : ordered moment; P_c : critical pressure to suppress T_N to $T = 0$.

¹ E-mail:jdt@lanl.gov

	T_c (K)	$\Delta C/\gamma_n T_c$	γ_n (J/mol K ²)	$H_{c2}^{a,b}$ (T)	H_{c2}^c (T)	$\partial H_{c2}^{a,b}/\partial T$ (T/K)	$\partial H_{c2}^c/\partial T$ (T/K)	$\xi_0^{a,b}$ (Å)	ξ_0^c (Å)
CeIrIn ₅	0.4 [4]	0.76 [4]	0.70 [4]	1.0 [11]	0.49 [11]	-4.8 [4]	-2.54 [20]	260 [11]	180 [11]
CeCoIn ₅	2.3 [3]	4.5 [3]	0.35 [3]	11.9 [21] 11.6 [22]	4.95 [21,22]	-24.0 [22]	-8.2 [20] -11.0 [22]	82 [22]	53 [22]
CeRhIn ₅ @ 2.1 GPa	2.12 [23]	0.36 [23]	0.38 [23]		10.2 [24] at 2.5 GPa		-15.0 [24] at 2.5 GPa	57 [24]	
Ce ₂ RhIn ₈ @ 1.63 GPa	1.1 [5]		≈ 0.20 [5]	5.4 [5]		-9.2 [5]			77 [5]

Table 2

Superconducting properties of $\text{Ce}_m\text{T}_n\text{In}_{3m+2n}$. T_c : superconducting transition temperature; $\Delta C/\gamma_n T_c$: jump in specific heat at T_c normalized by the Sommerfeld coefficient γ_n at $T \geq T_c$; $H_{c2}^{a,b}$ (H_{c2}^c): upper critical field in the a - b plane (parallel to the c -axis) extrapolated to $T = 0$; $\partial H_{c2}/\partial T$: slope of the upper critical field near T_c ; ξ_0 : Ginzburg-Landau superconducting coherence length at $T = 0$.

erties of these three compounds are summarized in table 1. The commensurate ordering Q -vector, ordered moment and P_c are similar in CeIn₃ and Ce₂RhIn₈; however, at 1.65 K, Ce₂RhIn₈ also develops an incommensurate magnetic structure [17], as does CeRhIn₅. From this comparison, the $n = 2$ member superficially appears to be a magnetic hybrid of the $n = 1$ and $n = \infty$ members, as might be expected from its crystal structure. Inelastic neutron scattering studies [18] of CeRhIn₅ find that magnetic correlations develop on a temperature scale roughly twice T_N . The correlation length along the tetragonal c -axis $\xi_c \approx 1.3$ c; whereas, in the a - b plane, the correlation length $\xi_a \approx 5$ a, reflecting magnetic anisotropy that may be important for superconductivity. Presently, we do not know if this anisotropy changes as the antiferromagnetic-superconducting boundary is approached with applied pressure, but it appears [19] that the c -axis discommensuration δ is somewhat pressure dependent.

A rather remarkable characteristic of this family of materials is their instability to superconductivity. Besides CeIn₃, CeRhIn₅ and Ce₂RhIn₈ under pressure, CeIrIn₅ and CeCoIn₅ are superconducting at atmospheric pressure. See table 2. In each case, superconductivity develops out of a highly correlated state with a large specific heat Sommerfeld coefficient γ_n and in proximity to antiferromagnetism. For example, substituting a small amount of Rh into CeCo_{1-x}Rh_xIn₅ or CeIr_{1-x}Rh_xIn₅ induces antiferromagnetism, and, for a range of x (roughly $0.3 \lesssim x \lesssim 0.7$), homogeneous coexistence of superconductivity and antiferromagnetism. [25,26,27] The superconducting transition temperatures also are high compared to other heavy-fermion examples. With applied pressure, all compounds listed in table 2 have T_c s between 2.2 and 2.6 K [5,24,28], except CeIrIn₅ whose bulk T_c reaches ≈ 1 K at 2.1 GPa [29,30] and does not exceed 1.2 K at pressures to 4 GPa [31].

Electronic anisotropy is reflected in an upper criti-

cal magnetic field that is typically two times larger for $H \parallel a - b$ plane than for $H \parallel c$ -axis. In many cases, the measured $H_{c2}(0)$ exceeds the Pauli paramagnetic limit $H_P/T_c = 1.86$ T/K. [32] In this regard, CeCoIn₅ has been studied most extensively and, for $H \parallel [001]$, exhibits a first order phase transition in a narrow field range at low temperatures [33], which is attributed to Pauli limiting. For $H \parallel [110]$, a magnetically hysteretic transition develops below $0.6T_c$ that is consistent with the formation of a spatially inhomogeneous Fulde-Ferrel-Larkin-Ovchinnikov state. [21] This possibility deserves further study. Additionally, $H_{c2}(0)$ is weakly, but clearly, anisotropic within the basal plane [21,34], implying the possibility of non-s-wave pairing.

There is growing evidence, summarized in table 3, that superconductivity in members of the family is unconventional. Power laws found deep in the superconducting state, $C/T \propto T$, thermal conductivity $\kappa \propto T^3$ and spin-lattice relaxation rate $1/T_1 \propto T^3$, are those expected of an order parameter with line nodes. Together with Knight shift measurements [36,42] on CeCoIn₅, these power laws suggest unconventional spin-singlet superconductivity, and, indeed, thermal conductivity measurements [34] find a prominent four-fold modulation in κ as a magnetic field is rotated in

	C/T	κ	$1/T_1$	λ
CeIrIn ₅	T [20]	T^3 [20]	T^3 [35,36]	$T^{1.5 \pm 0.2}$ [37]
CeCoIn ₅	T [20]	$T^{3.37}$ [20]	$T^{3+\epsilon}$ [36]	$T^{1.65 \pm 0.2}$ [37] $T^{1+\epsilon}$ [38] $T^{1.5}$ [39] $T^{\frac{3}{2}}/T$ [40]
CeRhIn ₅ @ 2.1 GPa	T [23]		T^3 [41]	

Table 3

Power laws in the superconducting state. C/T : specific heat divided by temperature; κ : electronic thermal conductivity; $1/T_1$: spin-lattice relaxation rate; λ : superconducting penetration depth.

	C/T	ρ	$1/T_1 T$
CeIn ₃		$T^{1.5-1.6}$ [12] near P_c	const. [43] $P \geq P_c$
CeIrIn ₅	$\gamma_0 - AT^{\frac{1}{2}}$ [44] for $H = 6$ T	$T^{1.3}$ [4]	$(T+8)^{-\frac{3}{4}}$ [35] $(T+0.86)^{-\frac{1}{2}}$ [36]
CeCoIn ₅	$-\ln T$ [3,45]	$T^{1.0 \pm 0.1}$ [28,45]	$\sim T^{-\frac{3}{4}}$ [36]
CeRhIn ₅		T^1 [24] $P = 3.2$ GPa	$(T+1.5)^{-\frac{1}{2}}$ [41] $P = 2.1$ GPa
Ce ₂ RhIn ₈		$T^{0.95 \pm 0.05}$ [5] $P = 1.63$ GPa	

Table 4

Non-Fermi-liquid behaviors. C/T : specific heat divided by temperature; ρ : electrical resistivity in the $a-b$ plane; $1/T_1 T$: spin-lattice relaxation rate divided by temperature.

the basal plane. The magnitude and location of maxima in $\kappa(\theta)$ are consistent with an order parameter having $d_{x^2-y^2}$ symmetry.

The boson mediating Cooper pairing remains unknown, but the preponderance of evidence points to antiferromagnetic spin fluctuations. The temperature dependence of some normal state properties further suggests that these fluctuations may not be conventional. For a Landau Fermi liquid, $C/T \sim \text{constant}$, $\rho \propto T^2$, and $1/T_1 T \sim \text{constant}$ are expected at low temperatures. As shown in table 4, this is not the case for several family members. These distinctly non-Fermi-liquid behaviors are expected [46] near an antiferromagnetic quantum-critical point and are found for the examples in table 4 only in $T-P-H$ space close to superconductivity. The functional dependencies, particularly $\rho \propto T$, suggest 2-dimensional antiferromagnetic quantum fluctuations. Understanding the interplay of electronic and magnetic anisotropies with quantum-critical fluctuations and superconductivity is one problem posed by this interesting family of heavy-fermion compounds.

Finally, we note the possible existence of a spin pseudogap in CeRhIn₅ near its critical pressure P_c [43] and in CeCoIn₅ for $0 \leq P \lesssim 1.6$ GPa [28]. The small difference in temperature scale (~ 5 K and ~ 3 K, respectively) on which a pseudogap signature appears in these two compounds seems to be related to their relative cell volumes. [28] An analysis of systematic changes in thermodynamic and transport properties of Ce_{1-x}La_xCoIn₅ further suggests a connection between the possible pseudogap in CeCoIn₅ and the development of short-range antiferromagnetic correlations. [45]

Acknowledgements Work at Los Alamos was performed under the auspices of the U.S. DOE Office of

Science. ZF acknowledges support by NSF grant DMR-9971348. We also thank V. A. Sidorov and H. A. Borges for communicating results of their unpublished pressure measurements.

References

- [1] H. Hegger *et al.*, Phys. Rev. Lett. **84** (2001) 4986.
- [2] J. D. Thompson *et al.*, J. Magn. Magn. Mat. **226** (2001) 5.
- [3] C. Petrovic *et al.*, J. Phys. Condens. Matter **13** (2001) L337.
- [4] C. Petrovic *et al.*, Europhys. Lett. **53** (2001) 354.
- [5] M. Nicklas *et al.*, cond-mat/0204064.
- [6] Yu N. Grin *et al.*, Sov. Phys. Crystallogr. **24** (1979) 137.
- [7] E. Moshopoulou *et al.*, J. Solid State Chem. **158** (2001) 25.
- [8] D. Hall *et al.*, Phys. Rev. B **64** (2001) 4506; D. Hall *et al.*, Phys. Rev. B **64** (2001) 212508.
- [9] Y. Haga *et al.*, Phys. Rev. B **63** (2001) 503.
- [10] R. Settai *et al.*, J. Phys. Condens. Matter **13** (2001) L627.
- [11] H. Shishido *et al.*, J. Phys. Soc. Jpn. **71** (2002) 162.
- [12] I. R. Walker *et al.*, Physica C **282-287** (1997) 3030; N. Mathur *et al.*, Nature **394** (1998) 39; G. Knebel *et al.*, Phys. Rev. B **65** (2001) 02225.
- [13] J. M. Lawrence and S. Shapiro, Phys. Rev. B **22** (1980) 4379.
- [14] A. Benoit *et al.*, Solid State Commun. **34** (1980) 293.
- [15] W. Bao *et al.*, Phys. Rev. B **62** (2001) 14621; Phys. Rev. B **63** (2001) 219901.
- [16] W. Bao *et al.*, Phys. Rev. B **64** (2001) 020401.
- [17] A. Malinowski *et al.*, unpublished; W. Bao *et al.*, unpublished.
- [18] W. Bao *et al.*, Phys. Rev. B **65** (2002) 100505.
- [19] A. Llobet *et al.*, unpublished.
- [20] R. Movshovich *et al.*, Phys. Rev. Lett. **86** (2001) 5152.
- [21] T. P. Murphy *et al.*, Phys. Rev. B **65** (2002) 100514.
- [22] S. Ikeda *et al.*, J. Phys. Soc. Jpn. **70** (2001) 2248.
- [23] R. A. Fisher *et al.*, Phys. Rev. B **65** (2002) 224509.
- [24] T. Muramatsu *et al.*, J. Phys. Soc. Jpn. **70** (2001) 3362.
- [25] P. G. Pagliuso *et al.*, Phys. Rev. B **64** (2001) 100503; P. G. Pagliuso *et al.*, cond-mat/0107266v2.
- [26] V. S. Zapf *et al.*, Phys. Rev. B **65** (2001) 014506.
- [27] G.-q. Zheng *et al.*, unpublished.
- [28] V. A. Sidorov *et al.*, cond-mat/0202251.
- [29] M. Nicklas *et al.*, unpublished.
- [30] R. Borth *et al.*, unpublished; G. Sparn *et al.*, unpublished.
- [31] V. A. Sidorov *et al.*, unpublished.
- [32] A. M. Clogston, Phys. Rev. Lett. **2** (1962) 9.
- [33] A. Bianchi *et al.*, cond-mat/0203310.
- [34] K. Izawa *et al.*, Phys. Rev. Lett. **87** (2001) 057002.

- [35] G.-q. Zheng *et al.*, Phys. Rev. Lett. **86** (2001) 4664.
- [36] Y. Kohori *et al.*, Phys. Rev. B **64** (2001) 134526.
- [37] W. Higimoto *et al.*, J. Phys. Soc. Jpn. **71** (2002) 1023.
- [38] R. J. Ormeno *et al.*, Phys. Rev. Lett. **88** (2002) 047005.
- [39] S. Özcan *et al.*, cond-mat/0206069.
- [40] E. Chia *et al.*, cond-mat/0206468.
- [41] T. Mito *et al.*, Phys. Rev. B **63** (2001) 220507.
- [42] N. J. Curro *et al.*, Phys. Rev. B **64** (2001) 18051.
- [43] S. Kawasaki *et al.*, Phys. Rev. B **65** (2001) 020504.
- [44] J. S. Kim *et al.*, Phys. Rev. B **64** (2001) 134524.
- [45] S. Nakatsuji *et al.*, cond-mat/0205496.
- [46] See, for example, G. R. Stewart, Rev. Mod. Phys. **73** (2001) 797.